

5 DIELECTRIC PORCELAIN COMPOSITION AND
DIELECTRIC RESONATOR USING THE COMPOSITION

 The present invention relates to a dielectric porcelain composition particularly excellent in characteristics at a millimeter-wave bandwidth region, a
10 dielectric resonator using the composition, and a process for the manufacture of the composition and resonator capable of controlling the characteristics (relative permittivity ϵ_r and temperature coefficient τ_f).

Description of the Prior Art:

 While various dielectric materials have been known as dielectric
15 materials for high frequency, magnesium titanate-based dielectric materials have been known as one of the materials having a relatively high Q_f value. According to "Ceramics Engineering Handbook," edited by Japan Ceramics Society and published by Gihodo, Vol. 1, p. 1885, May 30, 1993, MgTiO_3 that is a magnesium titanate-based dielectric material, has relative permittivity ϵ_r of 17, a
20 Q_f value of 110000 GHz and temperature dependency of resonance frequency (temperature coefficient τ_f) of -45 ppm/K.

 In addition, improvements in magnesium titanate-based materials have also been proposed. For example, JP-B SHO 61-14605 discloses a dielectric material obtained by sintering a material containing titanium dioxide and more
25 than 1 mole and not more than 1.3 moles of magnesium oxide per mole of the titanium dioxide. As the characteristics of the dielectric material of the prior art, it is disclosed that the relative permittivity $\epsilon_r = 17.3$ and no load $Q_u = 12000$ (120000 GHz in terms of the Q_f value) when $\text{MgO}:\text{TiO}_2 = 1.2:1$.

 JP-A 2002-193662 discloses dielectric porcelain comprising a first
30 crystal phase of at least one species consisting of MgTiO_3 , CaTiO_3 , Mg_2SiO_4 and BaTi_4O_9 , a second crystal phase of at least one species consisting of Mg_2TiO_4 ,

Mg₂B₂O₅ and Li₂TiSiO₅ and oxides of Si, B and Li, with the aim of materializing dielectric porcelain having a high Q value, with neither flexion nor distortion produced when being calcined together with a conductive material.

Though the technologies in the field of date communications have
5 recently been developed conspicuously, the characteristics required for dielectric materials used for dielectric resonators or other such devices, including the aforementioned Qf value, tend to be diversified due to applications, frequency bandwidths and the like.

In consideration of an application particularly as a resonator material,
10 it is required from a standpoint of ready design to develop dielectric materials having relative permittivity ϵ_r low to a certain extent as one of the characteristics of dielectric materials for submillimeter-wave and millimeter-wave regions. Since the dimension of a resonance phenomenon is directly proportionate to $\epsilon^{-1/2}$ when the dielectric constant is defined as ϵ , when a material having high relative
15 permittivity is used, the dimension of a resonator has to be extremely small with an increasing frequency. In order to make it easy to design a resonator, it is demanded to develop a dielectric material having appropriate relative permittivity ϵ_r taking the entire dimension and workability into consideration.

When the dielectric material is used for a resonator, it is generally
20 noted that the temperature coefficient τ_f is desirably as small as possible. It is further preferable in view of the temperature coefficient of the peripheral parts and other such surrounding parts that the temperature coefficient be set at an optional value to a certain extent.

From these viewpoints, the prior art technologies, such as those
25 disclosed by JP-B SHO 61-14605 and JP-A 2002-193662, for example, pay principal attention to an improvement in the Qf value and Q value, with relative permittivity ϵ_r and temperature coefficient τ_f taken little into consideration.

In the materials available on the market, which have small relative permittivity ϵ_r and small temperature coefficient τ_f , the former is about 12.6 and the latter is about -10 ppm/K. These values do not necessarily suffice.

The present invention has been proposed in view of the state of the
5 conventional affairs.

One object of the present invention is to provide a dielectric porcelain composition and a dielectric resonator using the composition, in which the relative permittivity ϵ_r can be adjusted to a relatively small value, and it is made possible to readily design submillimeter-wave resonators and millimeter-wave
10 resonators, for example.

Another object of the present invention is to provide a dielectric porcelain composition and a dielectric resonator using the composition, in which the temperature coefficient τ_f can be made small as much as possible and slightly adjusted in accordance with the surrounding circumstances and the like.

15 Still another object of the present invention is to provide a dielectric porcelain composition and a dielectric resonator using the composition, in which the relative permittivity ϵ_r has been adjusted to a small value to a certain extent and the temperature coefficient τ_f has been adjusted to the vicinity of zero.

Yet another object of the present invention is to provide a process for
20 optionally adjusting the characteristics (relative permittivity ϵ_r and temperature coefficient τ_f) of a dielectric porcelain composition.

The present inventors have keenly continued their studies over a long period of time in order to attain the objects mentioned above. As a result, they have found that addition of Mg_2SiO_4 to MgTiO_3 enables the relative permittivity
25 ϵ_r to be freely adjusted in accordance with the content of Mg_2SiO_4 , with the temperature coefficient τ_f varied little, and also to be set optimum in submillimeter-wave or millimeter-wave bandwidth regions and that addition of CaTiO_3 to MgTiO_3 enables the temperature coefficient τ_f to be set optional in the vicinity of zero in accordance with the content of CaTiO_3 , with the relative

permittivity ϵ_r not so much varied. The present invention has been perfected based on these findings.

According to one aspect of the present invention, a dielectric porcelain composition comprising MgTiO_3 and Mg_2SiO_4 is characterised in that the composition satisfies $a + b = 1$ and $0 < b < 1$, wherein a denotes a molar ratio of MgTiO_3 and b denotes a molar ratio of Mg_2SiO_4 ; a dielectric porcelain composition comprising MgTiO_3 and CaTiO_3 is characterised in that the composition satisfies $a + c = 1$ and $0 < c \leq 0.15$, wherein a denotes a molar ratio of MgTiO_3 and c denotes a molar ratio of CaTiO_3 ; or a dielectric porcelain composition comprising MgTiO_3 , Mg_2SiO_4 and CaTiO_3 is characterised in that the composition satisfies $a + b + c = 1$, $0 < b < 1$ and $0 < c \leq 0.15$, wherein a denotes a molar ratio of MgTiO_3 , b denotes a molar ratio of Mg_2SiO_4 and c denotes a molar ratio of CaTiO_3 .

In the dielectric porcelain compositions, relative permittivity ϵ_r and temperature coefficient τ_f can be obtained at optional values, respectively, in the range of 6.8 to 18 and in the range of -55 to $+55$ ppm/K. There can be realized a dielectric porcelain composition having relative permittivity ϵ_r in the vicinity of 10 and temperature coefficient τ_f in the vicinity of zero, for example.

The dielectric porcelain composition can be used as a dielectric material for dielectric resonators, such as submillimeter-wave resonators and millimeter-wave resonators. Therefore, the dielectric resonator of the present invention uses the dielectric porcelain composition as its resonator material.

As described above, the addition of Mg_2SiO_4 to MgTiO_3 enables the relative permittivity ϵ_r to be freely adjusted in accordance with the content of Mg_2SiO_4 , and the addition of CaTiO_3 to MgTiO_3 enables the temperature coefficient τ_f to be set optional in the vicinity of zero in accordance with the content of CaTiO_3 . In view of these, the adjustment of the contents of these components enables adjustment of the characteristics of a dielectric porcelain composition to be obtained.

According to another aspect of the present invention, there is provided a manufacturing process for the dielectric porcelain composition, which can control the characteristics of the composition. Specifically, the contents of Mg_2SiO_4 and CaTiO_3 are adjusted respectively within predetermined ranges to adjust the relative permittivity ϵ_r and temperature coefficient τ_f .

The above and other objects, characteristic features and advantages of the present invention will become apparent to those skilled in the art from the description to be give herein below with reference to the accompanying drawings, in which:

Figure 1 is a flow chart showing one example of a manufacturing process for a dielectric porcelain composition according to the present invention,

Figure 2 is an X-ray diffraction chart of $0.6\text{MgTiO}_3\text{-}0.4\text{Mg}_2\text{SiO}_4$,

Figure 3 is a characteristic diagram showing the relationship between Mg_2SiO_4 content and the relative permittivity ϵ_r in a system of $\text{MgTiO}_3\text{-Mg}_2\text{SiO}_4$,

Figure 4 is a characteristic diagram showing the relationship between Mg_2SiO_4 content and the temperature coefficient τ_f in the $\text{MgTiO}_3\text{-Mg}_2\text{SiO}_4$ system,

Figure 5 is a characteristic diagram showing the relationship between the calcining temperature and the relative density in the $\text{MgTiO}_3\text{-Mg}_2\text{SiO}_4$ system,

Figure 6 is an X-ray diffraction chart of $0.91\text{MgTiO}_3\text{-}0.09\text{CaTiO}_3$,

Figure 7 is a characteristic diagram showing the relationship between CaTiO_3 content and the relative permittivity ϵ_r in a system of $\text{MgTiO}_3\text{-CaTiO}_3$,

Figure 8 is a characteristic diagram showing the relationship between CaTiO_3 content and the temperature coefficient τ_f in the MgTiO_3 - CaTiO_3 system,

Figure 9 is a characteristic diagram showing the relationship between the calcining temperature and the relative density in the MgTiO_3 - CaTiO_3 system,

5 Figure 10 is an X-ray diffraction chart of 0.2275MgTiO_3 - $0.6825\text{Mg}_2\text{SiO}_4$ - 0.09CaTiO_3 ,

Figure 11 is a characteristic diagram showing the results of relative permittivity ϵ_r measured in a system of MgTiO_3 - Mg_2SiO_4 - CaTiO_3 , with 0.05 mol of CaTiO_3 fixed and with the Mg_2SiO_4 substitution amount varied,

10 Figure 12 is a characteristic diagram showing the results of temperature coefficient τ_f measured in the MgTiO_3 - Mg_2SiO_4 - CaTiO_3 system, with 0.05 mol of CaTiO_3 fixed and with the Mg_2SiO_4 substitution amount varied,

Figure 13 is a characteristic diagram showing the results of relative density measured in the MgTiO_3 - Mg_2SiO_4 - CaTiO_3 system, with 0.05 mol of CaTiO_3 fixed and with the Mg_2SiO_4 substitution amount varied,

15 Figure 14 is a characteristic diagram showing the results of relative permittivity ϵ_r measured in a system of MgTiO_3 - Mg_2SiO_4 - CaTiO_3 , with MgTiO_3 : Mg_2SiO_4 fixed to 1:3 and with the CaTiO_3 substitution amount varied,

Figure 15 is a characteristic diagram showing the results of temperature coefficient τ_f measured in the MgTiO_3 - Mg_2SiO_4 - CaTiO_3 system, with MgTiO_3 : Mg_2SiO_4 fixed to 1:3 and with the CaTiO_3 substitution amount varied, and

20 Figure 16 is a characteristic diagram showing the results of relative density measured in the MgTiO_3 - Mg_2SiO_4 - CaTiO_3 system, with MgTiO_3 : Mg_2SiO_4 fixed to 1:3 and with the CaTiO_3 substitution amount varied.

The dielectric porcelain composition, dielectric resonator using the composition and manufacturing process for the composition according to the present invention will be described hereinafter in detail.

The dielectric porcelain composition of the present invention

comprises magnesium titanate MaTiO_3 added with either one or both of Mg_2SiO_4 and CaTiO_3 .

Though MaTiO_3 has excellent characteristics that include a high Qf value, it exhibits slightly high relative permittivity ϵ_r of about 18.2 and large
5 temperature coefficient τ_f of -57 ppm/K. In view of this, added to MaTiO_3 in the present invention are Mg_2SiO_4 to improve the relative permittivity ϵ_r and CaTiO_3 to improve the temperature coefficient τ_f .

When Mg_2SiO_4 is added to MaTiO_3 , the relative permittivity ϵ_r decreases substantially in proportion to the content of Mg_2SiO_4 , whereas the
10 temperature coefficient τ_f varies little. On the other hand, when CaTiO_3 is added to MaTiO_3 , the temperature coefficient τ_f shifts gradually from the minus side to the plus side, whereas the relative permittivity ϵ_r arises little. For these reasons, the relative permittivity ϵ_r and temperature coefficient τ_f can independently be controlled depending on the contents of Mg_2SiO_4 and CaTiO_3 .

15 From these standpoints, Mg_2SiO_4 or CaTiO_3 is added to MaTiO_3 . It is preferable that the content of Mg_2SiO_4 should satisfy that $a + b = 1$ and that $0 < b < 1$, wherein a denotes the molar ratio of MgTiO_3 and b denotes the molar ratio of Mg_2SiO_4 . Controlling the content of Mg_2SiO_4 optionally to satisfy these enables the relative permittivity ϵ_r to be freely controlled to a value lower than
20 that MaTiO_3 has, e.g. in the range of 6.8 to 18. However, when the relative permittivity ϵ_r is to be set at a value suitable for use in submillimeter-wave and millimeter-wave regions, e.g. a value of not more than 12, the b is more preferably defined as $0.5 \leq b < 1$.

In the case of CaTiO_3 added to MgTiO_3 , it is preferable that the content thereof should satisfy that $a + c = 1$ and that $0 < c \leq 0.15$, wherein a denotes a molar ratio of MgTiO_3 and c denotes a molar ratio of CaTiO_3 . Controlling the content of CaTiO_3 optionally to satisfy these enables the temperature coefficient τ_f to be freely controlled to a value in the range of -55 to $+55$ ppm/K. In order to control the temperature coefficient τ_f to be as close as zero, i.e. around 30 ppm/K, however, the c is more preferably defined as $0.03 \leq c \leq 0.08$.

When both Mg_2SiO_4 and CaTiO_3 are added to MgTiO_3 , the contents thereof may be adjusted to respectively suitable amounts to satisfy that $a + b + c = 1$, that $0 < b < 1$ and that $0 < c \leq 0.15$, wherein a denotes a molar ratio of MgTiO_3 , b denotes a molar ratio of Mg_2SiO_4 and c denotes a molar ratio of CaTiO_3 .

In order to control the relative permittivity ϵ_r to be a value suitable for use in submillimeter-wave and millimeter-wave regions, e.g. a value of not more than 12, and the temperature coefficient τ_f to be as close as zero, the contents of Mg_2SiO_4 and CaTiO_3 may be adjusted as mentioned above. Though their respective optimum values are slightly different from the defined values, the more preferably ranges are $0.5 \leq b < 1$ and $0.05 \leq c \leq 0.09$, respectively.

It is noted that since it is clear from the X-ray diffraction that the respective components of the dielectric porcelain composition exist respectively in the form of MgTiO_3 , Mg_2SiO_4 and CaTiO_3 and that the matrix thereof is a crystal phase having the three components combined, the composition is to be represented by their ratios in mol.

Based on the above, by controlling the ratios of the respective components, a dielectric porcelain composition exhibiting the relative permittivity ϵ_r of 10.86, temperature coefficient τ_f of -2.7 ppm/K and Q_f value of 74000 GHz can be materialised.

The manufacturing process for the dielectric porcelain composition according to the present invention will be described herein below. The flow chart thereof adopted by the present invention is as shown in Figure 1.

In the manufacturing process of the present invention, MgO, TiO₂,
5 CaCO₃ and SiO₂ are used as raw materials, for example. While the respective components are mixed in accordance with their respectively desired characteristics, since the prepared composition is reflected substantially as it is by the composition of the dielectric porcelain composition, the raw material components are mixed so that the prepared composition and the composition of
10 the dielectric porcelain composition can have the relationship of 1:1.

The process of manufacturing the dielectric porcelain composition will be described. The raw materials, MgO, TiO₂, CaCO₃ and SiO₂, are mixed at a mixing process 1 to obtain a mixture. The mixture is subjected to a drying process 2 and a shaping process 3 and preliminarily calcined at a calcining
15 process 4. The preliminary calcining is performed in order for the reaction of the raw materials to proceed to a certain extent and generally at a temperature slightly lower than that used in the sintering.

The preliminarily calcined product is milled at a milling process 5 and then dried at a drying process 6. The dried product is granulated at a
20 granulating process 7. In the granulating process, a binder is mingled with the dried product. Though any optional binder can be used, polyvinyl alcohol or the like can advantageously be used, for example.

The granulated product is shaped into a desired shape at a shaping process 8 and sintered at a sintering process 9. The sintering temperature used
25 at the sintering process is adjusted in the range of 1250°C to 1500°C, for example. The optimum sintering temperature is made slightly different depending on the raw materials for the dielectric porcelain composition. When manufacturing a dielectric porcelain composition comprising MgTiO₃ and Mg₂SiO₄, the sintering temperature of not less than 1300°C is preferred. In this case, when the sintering

temperature is less than 1300°C, the Qf value will be lowered and the relative density will also be lowered. In the case of a dielectric porcelain composition comprising MgTiO_3 and CaTiO_3 , it is preferable to use the sintering temperature of not less than 1250°C. When it is less than 1250°C, both the Qf value and the
5 relative density are lowered similarly to the case mentioned above. When a dielectric porcelain composition comprising MgTiO_3 , Mg_2SiO_4 and CaTiO_3 is to be manufactured, the sintering temperature of not less than 1300°C is preferable. By setting the sintering temperature within the aforementioned range, the Qf value and relative density can be maintained at high levels, respectively.

10 In the manufacturing process, MgO , TiO_2 , CaCO_3 and SiO_2 are used as the raw materials. However, this is by no means limitative. For example, MgTiO_3 , Mg_2SiO_4 and CaTiO_3 can be prepared in advance at their predetermined ratios and used in the manufacturing process.

The dielectric porcelain composition can be used at the frequency
15 bandwidths of submillimeter-waves and millimeter-waves, e.g. 30 to 300 GHz. The frequency bandwidths include that of a radar for automobiles (using the frequency of 77GHz:38.5GHz multiplied by 2).

Therefore, the dielectric porcelain composition of the present invention can be used as a material for resonators used in the submillimeter-wave
20 and millimeter-wave regions and a substrate material for MIC dielectrics, and for dielectric waveguides, dielectric antennas, impedance matching of various kinds of millimeter-wave circuits and other such electronic parts. It can suitably be used for dielectric resonators.

The present invention will be described based on concrete
25 experimental results.

Samples of a dielectric porcelain composition were produced in accordance with the following procedure.

MgO , TiO_2 , CaCO_3 and SiO_2 were weighed so that these raw materials had a predetermined mixing ratio and then mixed with a ball mill for 16 hours.

The mixture obtained was dried at 120°C for 24 hours and then shaped under a shaping pressure of 200 kgf/cm² into a disc 60 mm in diameter.

The disc was preliminarily calcined at 1100°C for 2 hours, then milled for 16 hours using the ball mill and dried at 120°C for 24 hours. The dried
5 product was granulated, with 1% by weight of polyvinyl alcohol added thereto, and shaped under a shaping pressure of 2000 kgf/cm² into a 12 mm-diameter.

Finally, the shaped product was principally calcined to obtain dielectric porcelain composition samples.

In accordance with the process of producing the dielectric porcelain
10 composition samples, MgTiO₃ and Mg₂SiO₄ used as raw materials were mixed so that b is in the range of 0 to 1, provided that $a + b = 1$, when the molar ratio of MgTiO₃ was defined as a and the molar ratio of Mg₂SiO₄ was defined as b . The mixture was sintered at a temperature of 1250°C to 1500°C to obtain various samples.

15 A sample of 0.6MgTiO₃-0.4Mg₂SiO₄, wherein $a = 0.6$ and $b = 0.4$, was measured using an X-ray diffraction apparatus. The results of measurement are shown in Figure 2. It can be observed from the X-ray diffraction chart that there exist peaks resulting respectively from MgTiO₃ and Mg₂SiO₄, from which it is found that the sample comprises a mixed crystal of MgTiO₃ and Mg₂SiO₄.

20 The relative permittivity ϵ_r and temperature coefficient τ_f of each sample were measured in accordance with the "Method of Testing Dielectric Characteristics of Fine Ceramics for Microwaves" of the Japanese Industrial Standards (JIS R 1627). The results of relative permittivity ϵ_r measurement are shown in Figure 3 and Table 1 below, and the results of temperature coefficient τ_f
25 measurement are shown in Figure 4 and Table 2 below.

Table 1

Molar ratio b of Mg_2SiO_4	Relative permittivity ϵ_r at 1300°C	Relative permittivity ϵ_r at 1350°C	Relative permittivity ϵ_r at 1400°C	Relative permittivity ϵ_r at 1450°C	Relative permittivity ϵ_r at 1500°C
0.0	17.90	18.16	18.24	18.17	18.22
0.1	16.05	16.33	16.36	16.24	16.07
0.2	14.06	14.61	14.67	14.64	14.41
0.4	11.23	11.88	11.99	11.93	11.84
0.5	10.25	10.53	10.64	10.63	10.62
0.6	9.54	9.59	9.65	9.67	9.69
0.8	7.93	7.99	8.08	8.14	8.16
0.9	7.26	7.41	7.45	7.47	7.49
1.0	6.55	6.89	6.98	6.99	6.91

Table 2

Molar ratio b of Mg_2SiO_4	Temperature coefficient τ_f at 1450°C (ppm/K)
0.0	-57.6
0.1	-52.1
0.2	-58.0
0.4	-60.3
0.5	-62.1
0.6	-62.8
0.8	-63.1
0.9	-64.0
1.0	-65.3

5

As is clear from Figure 3 and Table 1 above, it is found that the relative permittivity ϵ_r gradually decreases in proportion as the Mg_2SiO_4 content increases. As shown in Figure 4 and Table 2 above, it is found that the temperature coefficient τ_f varies little even when the Mg_2SiO_4 content varies.

10 This means that controlling the Mg_2SiO_4 content enables the relative permittivity ϵ_r to be controlled without affecting the other characteristic (temperature coefficient τ_f). Particularly when the molar ratio b of Mg_2SiO_4 is set to be 0.5 or more, the relative permittivity ϵ_r of 12 or less can be materialised.

The relative density of each sample produced was also measured.

The results of measurement are shown in Figure 5 and Table 3 below. As is clear from Figure 5 and Table 3, while the relative density shows a slight drop at 1300°C, it varies little at a temperature of more than 1300°C. Desired relative density could not be obtained when the calcining temperature was 1200°C or less (not shown). Therefore, when Mg_2SiO_4 is used to control the relative permittivity ϵ_r , it can be said that the calcining temperature is preferably set to be 1300°C or more.

Table 3

Molar ratio b of Mg_2SiO_4	Relative density (%) at 1300°C	Relative density (%) at 1350°C	Relative density (%) at 1400°C	Relative density (%) at 1450°C	Relative density (%) at 1500°C
0.0	97.5	98.4	99.0	98.7	98.6
0.1	97.6	98.6	98.6	98.0	97.6
0.2	95.5	97.7	98.2	97.8	97.1
0.4	93.5	96.9	97.4	97.3	96.8
0.5	92.9	96.3	97.9	97.2	97.1
0.6	92.5	95.6	98.2	97.0	96.9
0.8	91.5	94.8	97.6	97.0	96.9
0.9	90.9	94.0	97.6	96.9	97.8
1.0	90.2	92.3	97.8	97.1	97.5

In accordance with the process of producing the dielectric porcelain composition samples, MgTiO_3 and CaTiO_3 used as raw materials were mixed so that c is in the range of 0 to 0.09, provided that $a + c = 1$, when the molar ratio of MgTiO_3 was defined as a and the molar ratio of CaTiO_3 was defined as c . The mixture was principally calcined at a temperature of 1300°C to obtain various samples.

In Figure 6, shown are measurement results of a sample of $0.91\text{MgTiO}_3\text{-}0.09\text{CaTiO}_3$, wherein $a = 0.91$ and $c = 0.09$, measured using an X-ray diffraction apparatus. It can be observed from the X-ray diffraction chart that there exist peaks resulting respectively from MgTiO_3 and CaTiO_3 , from which it is found that the sample comprises a mixed crystal of MgTiO_3 and CaTiO_3 .

The relative permittivity ϵ_r and temperature coefficient τ_f of each sample were measured in accordance with the "Method of Testing Dielectric Characteristics of Fine Ceramics for Microwaves" of the Japanese Industrial Standards (JIS R 1627). The results of relative permittivity ϵ_r measurement are shown in Figure 7 and Table 4 below, and the results of temperature coefficient τ_f measurement are shown in Figure 8 and Table 5 below.

Table 4

Molar ratio c of CaTiO_3	Relative permittivity ϵ_r at 1250°C	Relative permittivity ϵ_r at 1300°C	Relative permittivity ϵ_r at 1350°C	Relative permittivity ϵ_r at 1400°C	Relative permittivity ϵ_r at 1450°C
0.00	17.80	17.90	18.16	18.24	18.17
0.05	19.80	19.94	20.08	20.51	20.47
0.07	21.22	21.45	21.70	21.98	22.02
0.09	22.82	22.98	23.27	23.52	23.41

Table 5

Molar ratio c of CaTiO_3	Temperature coefficient τ_f at 1300°C (ppm/K)
0.00	-57.6
0.05	-8.6
0.07	18.7
0.09	48.3

10

As is clear from Figure 8 and Table 5 above, it is found that the temperature coefficient τ_f gradually varies in proportion as the CaTiO_3 content increases. When the molar ratio c of CaTiO_3 is around 0.06, the temperature coefficient τ_f becomes substantially zero, and shifts to the minus side when the ratio is smaller than 0.06 and to the plus side when the ratio is larger than 0.06. On the other hand, the relative permittivity ϵ_r does not vary so much even when the CaTiO_3 content varies, as shown in Figure 7 and Table 4 above. This means that controlling the CaTiO_3 content enables the temperature coefficient τ_f to be independently controlled. Particularly when the molar ratio c of CaTiO_3 is set to be in the range of 0.03 to 0.08, the temperature coefficient τ_f can be controlled

20

in the range of ± 30 ppm/K.

The relative density of each sample produced was also measured. The results of measurement are shown in Figure 9 and Table 6 below. As is clear from Figure 9 and Table 6, the relative density shows a level not giving rise to any problem when the temperature is 1250°C or more. Therefore, when CaTiO₃ is used to control the temperature coefficient τ_f , it can be said that the calcining temperature is preferably set to be 1250°C or more.

Table 6

Molar ratio c of CaTiO ₃	Relative density (%) at 1250°C	Relative density (%) at 1300°C	Relative density (%) at 1350°C	Relative density (%) at 1400°C	Relative density (%) at 1450°C
0.00	97.0	97.5	98.4	99.0	98.7
0.05	95.6	96.4	96.8	98.1	98.2
0.07	96.2	96.8	97.7	98.8	99.1
0.09	96.8	97.9	98.3	99.2	99.1

In accordance with the process of producing the dielectric porcelain composition samples, samples each comprising MgTiO₃, Mg₂SiO₄ and CaTiO₃ were produced.

Figure 10 shows measurement results of a sample of 0.2275MgTiO₃-0.6825Mg₂SiO₄-0.09CaTiO₃, wherein $a = 0.2275$, $b = 0.6825$ and $c = 0.09$, measured using an X-ray diffraction apparatus. It can be observed from the X-ray diffraction chart that there exist peaks resulting respectively from MgTiO₃, Mg₂SiO₄ and CaTiO₃, from which it is found that the sample comprises a mixed crystal of MgTiO₃, Mg₂SiO₄ and CaTiO₃.

In a system of MgTiO₃-Mg₂SiO₄-CaTiO₃, various samples were prepared, with 0.05 mol of CaTiO₃ fixed ($c = 0.05$) and with the Mg₂SiO₄ substitution amount varied. In this case, the results of relative permittivity ϵ_r measured are shown in Figure 11 and Table 7 below, the results of temperature coefficient τ_f measured in Figure 12 and Table 8 below, and the relative density measured in Figure 13 and Table 9 below.

Table 7

Molar ratio b of Mg_2SiO_4	Relative permittivity ϵ_r at 1300°C	Relative permittivity ϵ_r at 1350°C	Relative permittivity ϵ_r at 1400°C
0.0000	19.94	20.08	20.51
0.2375	15.41	15.24	14.57
0.4750	11.85	11.31	11.25
0.7125	9.46	9.42	9.43
0.9500	7.60	7.83	7.48

Table 8

Molar ratio b of Mg_2SiO_4	Temperature coefficient τ_f at 1300°C (ppm/K)
0.0000	-8.6
0.2375	-21.1
0.4750	-30.5
0.7125	-40.6
0.9500	-44.7

5

Table 9

Molar ratio b of Mg_2SiO_4	Relative density (%) at 1300°C	Relative density (%) at 1350°C	Relative density (%) at 1400°C
0.0000	96.4	96.8	98.1
0.2375	96.9	96.7	94.7
0.4750	96.2	93.8	92.3
0.7125	97.1	96.0	95.9
0.9500	96.1	98.3	96.5

In the $\text{MgTiO}_3\text{-Mg}_2\text{SiO}_4\text{-CaTiO}_3$ system, various samples were produced, with $\text{MgTiO}_3\text{:Mg}_2\text{SiO}_4$ fixed to 1:3 and with the CaTiO_3 substitution amount varied. In this case, the results of relative permittivity ϵ_r measured are shown in Figure 14 and Table 10 below, the results of temperature coefficient τ_f measured in Figure 15 and Table 11 below, and the relative density measured in Figure 16 and Table 12 below.

Table 10

Molar ratio c of CaTiO_3	Relative permittivity ϵ_r at 1300°C	Relative permittivity ϵ_r at 1350°C	Relative permittivity ϵ_r at 1400°C
0.00		8.43	8.45
0.05	9.46	9.42	9.43
0.07	10.01	9.91	9.95
0.09	10.70	10.86	10.84

Table 11

Molar ratio c of CaTiO_3	Temperature coefficient τ_f at 1350°C (ppm/K)
0.00	-62.0
0.05	-40.6
0.07	-27.3
0.09	-2.9

5

Table 12

Molar ratio c of CaTiO_3	Relative density (%) at 1300°C	Relative density (%) at 1350°C	Relative density (%) at 1400°C
0.00		95.8	96.7
0.05	97.1	96.0	95.9
0.07	96.5	95.8	95.6
0.09	95.5	97.4	96.6

As is clear from these Figures and Tables, also in the three-element system, it is possible to control the relative permittivity ϵ_r through adjustment of the Mg_2SiO_4 content and control the temperature coefficient τ_f through adjustment of the CaTiO_3 content. In the composition of $0.2275\text{MgTiO}_3-0.6825\text{Mg}_2\text{SiO}_4-0.09\text{CaTiO}_3$ prepared with the aim that the relative permittivity $\epsilon_r = 10$ and that the temperature coefficient $\tau_f = 0$, it was found that the relative permittivity $\epsilon_r = 10.86$ and that the temperature coefficient $\tau_f = -2.7$ ppm/K.

15

In addition, upon considering the relative density of each sample produced, it was found from Figures 13 and 16 and Tables 9 and 12 that good

results could be obtained when the calcining temperature was 1300°C or more.

As is clear from the foregoing description, according to the present invention, the relative permittivity ϵ_r and temperature coefficient τ_f can be controlled to enable provision of a dielectric porcelain composition with the
5 relative permittivity ϵ_r suitable for submillimeter-wave and millimeter-wave regions and the temperature coefficient τ_f controlled to a value in the vicinity of 0.

Also according to the present invention, using the dielectric porcelain composition as a dielectric material enables provision of a dielectric resonator usable in the submillimeter-wave and millimeter-wave bandwidth regions. In
10 the dielectric resonator, since the dielectric porcelain composition exhibits appropriate relative permittivity ϵ_r , the dimensional tolerance can be alleviated to make it easy to design a dielectric resonator when being fabricated. Furthermore, the temperature coefficient can also be controlled in compliance with the temperature coefficient of the surrounding parts and the like.

15 The present disclosure relates to subject matter contained in Japanese Patent Application Nos. 2003-071545, filed on March 17, 2003, the contents of which are herein expressly incorporated by reference in its entirety.